

Magnon Condensation and Spin Superfluidity

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Abstract

We consider the phenomenon of Bose-Einstein condensation of quasi-equilibrium magnons which leads to a spin superfluidity, the coherent quantum transfer of magnetization in magnetic materials. These phenomena are beyond the classical Landau-Lifshitz-Gilbert paradigm. The critical conditions for excited magnon density for ferro- and antiferromagnets, bulk and thin films are estimated and discussed. The BEC should occur in the antiferromagnetic hematite at much lower excited magnon density compared to the ferromagnetic YIG.

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INTRODUCTION

It is well known that deviations of spins from the magnetic order in magnetic materials (ferromagnets, antiferromagnets and ferrites) have a collective character and are described in terms of spin waves and their quanta, magnons. Magnons are quasiparticles which represent very useful and universal theoretical concept and tool to describe various dynamic and thermodynamic processes in magnets. For example, analyzing thermodynamics of magnon gas, we obtain complete picture of thermodynamic properties of magnetic system. Since magnons have a magnetic moment, one can create extra magnons by the external pumping, alternating magnetic field, and increase disorder in the magnetic system. However, in certain conditions, increase the density of magnons can lead to entirely new states be called magnon condensates, macroscopic number of magnons in coherent quantum states (see, e.g., [1], [2]). These macroscopic quantum states significantly alter the properties of the magnetic system, its dynamics and transport. For example, a single-particle long-range coherency occurs in a quasi-equilibrium as the phenomenon of Bose - Einstein condensation (BEC) of magnons on the bottom of their spectrum. And the spatial gradients of these states leads to the phenomenon of spin superfluidity - the coherent transport of deflected magnetization. The spin superfluidity is an extremely interesting and promising phenomenon both for fundamental and applied studies. The main feature of spin superfluidity is the transfer of magnetization in magnetic materials for long distances. The real superfluidity means a long distance correlation of a non-diagonal terms of the density matrix, which does not, in general, take place in magnetically ordered materials. The short distance correlation of non-diagonal terms appears due to spin waves, or, magnons.

The experimental evidence that magnon BEC is a process, in which the coherent magnetization precession emerges spontaneously was demonstrated in the experiments with superfluid antiferromagnetic liquid crystal $^3\text{He-B}$ in 1984 [3]. Note that the magnetic part of its hamiltonian does not related to superfluidity. In the experiments [3] the magnetisation was deflected by RF pulse on a big angle of order 90° . After the pulse the induction decay signal dephasing with the time scale T_2^* which determins by inhomogeneous broadening. Indeed, after a some delay the induction signal spontaneously restoring and ringing a time scale of few orders of magnitude longer then T_2^* . This spontaneously restoring of the signal corresponds well to the principal of magnon BEC. The theoretical explanation of this

phenomena was done in [4] on the basis of minimization of global Ginzburg-Landau energy potential, the same way as later was done for to explain the atomic BEC formation [5]. A bunch of complex spin-superfluid phenomena was demonstrated for antiferromagnetic $^3\text{He-B}$: transport of magnetization by spin supercurrent between two cells with magnon BEC; phase-slip processes at the critical current; spin current Josephson effect; spin current vortex formation; Goldstone modes of magnon BEC oscillations etc. The comprehensive reviews of these experiments may be found in [6–8].

Later magnon BEC states were observed in different magnetic systems: in antiferromagnetic superfluid $^3\text{He-A}$ [9, 10]; in in-plane magnetized yttrium iron garnet $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) film (two minima in the spin-wave spectrum) [11, 12]; in antiferromagnets MnCO_3 and CsMnF_3 with Suhl-Nakamura indirect nuclear spin-spin interaction [13–15]. Corresponding nuclear spin superfluidity were reported in Ref.[14].

A microscopic theory of magnon BEC was proposed and developed in Refs.[16]–[18] (KS theory). It was found that strong, either parallel or perpendicular pumping of magnons leads to rapid growth of magnons and saturation with quasi-equilibrium in magnon system with an effective temperature T and effective chemical potential μ . Arising out of the exchange interaction, magnon-magnon scattering processes retain the total number of quasiparticles, ”dressed” magnons in the system and hold their distribution function of the form

$$n_k = \frac{1}{\exp \frac{\varepsilon_{\mathbf{k}} - \mu}{k_B T} - 1}. \quad (1)$$

The instability at $\mu = \min \varepsilon_{\mathbf{k}}$ in the quasi-equilibrium magnetic system is an analog of Bose-Einstein condensation phenomenon for the bottom dressed magnons.

Initially, the KS theory gave a qualitative description of the magnon parallel pumping experiments ([19],[20] (YIG at room temperature) and [21] (nuclear magnons in CsMnF_3), where the accumulation of magnons at the bottom of the spin wave spectrum was observed. More than decade later, purposeful experiment [11] directly demonstrated BEC of quasiequilibrium magnons in the thin film of YIG. Subsequent experimental studies have shown qualitative correspondance with the predicted distribution of excited magnons [22] and agreement with BEC under noisy pumping [23].

Thus one can say that the phenomenon of Bose-Einstein condensation of quasi-equilibrium magnons is a fundamental law of magnetic physics. In the present time the

main paradigm of the magnetic dynamics is the classical Landau-Lifshitz-Gilbert equation that *do not contain* information about quantum magnon condensates. BEC is a principal result of quantum statistics of quasiparticles in magnetic systems and it can exist at room (and even higher) temperatures. A superfluid spin current, a coherent quantum carrier of magnetization, is a common feature of BEC.

In this paper we consider critical conditions of quasi-equilibrium magnons for BEC formation in ferro- and antiferromagnets, bulk and thin films.

BEC OF BOSE PARTICLES

Let us first briefly discuss BEC of real bose particles. Their Bose-Einstein distribution is defined by Eq.(1), where $\varepsilon_k = \hbar k^2/2m$ is the kinetic energy of particles with wave vektor k . One can write the total number of bosons in the system as

$$N(\mu, T) = V_s \int n_k \frac{d^3k}{(2\pi)^3} \quad (2)$$

Here V_s is the volume of the sample. Using condition of BEC $\mu = \min \varepsilon_k$, from (2) we can obtain well-known formula for the BEC temperature versus density of bosons:

$$T_{BEC} = \kappa_0 \frac{\hbar^2}{k_B m} \left(\frac{N}{V_s} \right)^{2/3}, \quad \kappa_0 = \frac{2\pi}{[\zeta(\frac{3}{2})]^{2/3}} \simeq 3.31. \quad (3)$$

It should be noted that Bose-Einstein condensation is formed by particles with high populations when

$$n_k \simeq \frac{k_B T}{\varepsilon_k - \mu}. \quad (4)$$

We can prove it by direct calculation. Substituting this high temperature population (4) to Eq.(2) and cut the integral upper limit by the thermal particle energy $\varepsilon_k \simeq k_B T$, one obtains:

$$T_{BEC} \simeq \tilde{\kappa}_0 \frac{\hbar^2}{k_B m} \left(\frac{N}{V_s} \right)^{2/3}, \quad \tilde{\kappa}_0 = \frac{\pi^{4/3}}{2^{1/3}} \simeq 3.65. \quad (5)$$

Note that the only difference between (3) and (5) is the numerical factor within 10% accuracy.

BEC OF MAGNONS

As it was mentioned above, we can consider a Bose-Einstein condensation of dressed magnons at $\mu = \min \varepsilon_k$, as an instability in the quasiequilibrium magnetic system pumped by the external sources. The energy of a dressed magnon is defined by $\varepsilon_k = \varepsilon_k^{(0)} + \delta\varepsilon_k$, where $\varepsilon_k^{(0)} = \hbar\omega_k$ is the energy spectrum of bare magnons and $\delta\varepsilon_k$ is the energy shift due to magnon nonlinearities. Usually, the energy shift is much less than the energy gap $\min \varepsilon_k$ in the spin-wave spectrum. Therefore we can use an approximation $\varepsilon_k \simeq \varepsilon_k^{(0)}$. In addition, the critical density $N(\mu = \min \varepsilon_k, T)/V_s$ of magnons contains both the density of thermal magnons $N(0, T)/V_s$ at a given temperature and the effective density of pumped magnons N_p/V_s .

BEC in a ferromagnet

Let us consider first a ferromagnetic system with the quadratic spectrum (we neglect details of dipole-dipole interactions):

$$\omega_k = \omega_0 + \omega_{ex} (ak)^2. \quad (6)$$

The quasiequilibrium BEC will be entirely determined by pumping if the density of pumped magnons is much greater than the thermal magnon density ($N_p \gg N(0, T)$). In this case we obtain an analog of Eq.(3):

$$T_{BEC} = \kappa_0 \frac{2\hbar\omega_{ex}}{k_B} \left(a^3 \frac{N_p}{V_s} \right)^{2/3}, \quad (7)$$

or,

$$T_{BEC} \simeq \tilde{\kappa}_0 \frac{2\hbar\omega_{ex}}{k_B} \left(a^3 \frac{N}{V_s} \right)^{2/3} \quad (8)$$

in the high-population approximation.

This obtained above formula, does not give a BEC estimate if $N_p \lesssim N(0, T)$. Using high-population approximation, we write

$$N_p \simeq V_s \int_{\omega_0}^{\omega_T} \left(\frac{k_B T}{\hbar\omega_k - \mu} - \frac{k_B T}{\hbar\omega_k} \right) \frac{k^2 dk}{2\pi^2}, \quad (9)$$

and obtain at $\mu = \hbar\omega_0$

$$\begin{aligned} \frac{N_p}{V_s} &\simeq \frac{k_B T_{BEC}}{4\pi\hbar a^3} \frac{\omega_0^{1/2}}{\omega_{ex}^{3/2}}, \\ T_{BEC} &\simeq 4\pi \frac{\hbar\omega_{ex}}{k_B} \left(\frac{\omega_{ex}}{\omega_0} \right)^{1/2} \left(a^3 \frac{N_p}{V_s} \right), \end{aligned} \quad (10)$$

which coincides (with the accuracy of notations) with the exact calculation in Ref.[2]. An estimate for YIG, where $\omega_{ex}a^2 = 0.092 \text{ cm}^{-2}\text{s}^{-1}$ for $\omega_0 = 2\pi \times 2.5 \text{ GHz}$ gives $T_{BEC} \simeq 2.14 \times 10^{-17} (N_p/V_s) \text{ cm}^3\text{K}$. Thus we obtain a room-temperature BEC $T_{BEC} \simeq 300 \text{ K}$ at the pumped magnon density $N_p/V_s = 1.41 \times 10^{19} \text{ cm}^{-3}$.

BEC in an antiferromagnet

Consider now the magnon spectrum of the form

$$\omega_k = \sqrt{\omega_0^2 + \omega_{ex}^2 (ak)^2}, \quad (11)$$

which is typical for magnons in the "easy-plane" (or, canted) antiferromagnets. Taking into account that

$$k = \frac{\sqrt{\omega_k^2 - \omega_0^2}}{a\omega_{ex}} \quad \text{and} \quad kdk = \frac{\omega_k d\omega_k}{\omega_{ex}^2 a^2},$$

one can write

$$\frac{N(\mu = \hbar\omega_0, T)}{V_s} \simeq \frac{k_B T}{2\pi^2 \hbar} \frac{1}{a^3 \omega_{ex}^3} \int_{\omega_0}^{\omega_T} \sqrt{\frac{\omega + \omega_0}{\omega - \omega_0}} \omega d\omega. \quad (12)$$

If $N_p \gg N(0, T)$, for $k_B T \gg \hbar\omega_0$ we have

$$T_{BEC} \simeq (2\pi)^{2/3} \frac{\hbar\omega_{ex}}{k_B} \left(a^3 \frac{N_p}{V_s} \right)^{1/3}. \quad (13)$$

If $N_p \lesssim N(0, T)$, one can rewrite Eq.(9) in the form:

$$\frac{N_p}{V_s} \simeq \frac{k_B T}{2\pi^2 \hbar} \frac{\omega_0}{a^3 \omega_{ex}^3} \int_{\omega_0}^{\omega_T} \sqrt{\frac{\omega + \omega_0}{\omega - \omega_0}} d\omega. \quad (14)$$

After integration, at $k_B T \gg \hbar\omega_0$ we obtain

$$\frac{N_p}{V_s} \simeq \frac{(k_B T_{BEC})^2}{2\pi^2 \hbar^2} \frac{\omega_0}{a^3 \omega_{ex}^3},$$

or,

$$T_{BEC} \simeq \sqrt{2\pi} \frac{\hbar\omega_{ex}}{k_B} \left(\frac{\omega_{ex}}{\omega_0} \right)^{1/2} \left(a^3 \frac{N_p}{V_s} \right)^{1/2}. \quad (15)$$

Note that the BEC temperature for antiferromagnet has lower power dependence on small parameter $a^3 N_p/V_s \ll 1$ and therefore one can expect much lower densities of pumped magnons to achieve condensation. An estimate for $\alpha\text{-Fe}_2\text{O}_3$ (hematite), where $\omega_{ex}a \approx 24 \times 10^5$ cm/s for $\omega_0 = 2\pi \times 2.5$ GHz gives $T_{BEC} \approx 10^{-6}(N_p/V_s)^{1/2}$ cm^{3/2}K. Thus we obtain a room-temperature BEC, $T = 300$ K at $N_p/V_s = 0.89 \times 10^{16}$ cm⁻³.

BEC STATE IN A FERROMAGNETIC FILM

Let us now consider an ultra-thin ferromagnetic film. There are two principal cases: 1) external magnetic field \mathbf{H} is parallel to the surface of the film and 2) \mathbf{H} is perpendicular to the film surface. In the first case, the BEC condition $\mu = \min \hbar\omega_{\mathbf{k}}$ give us two minima at $\pm \mathbf{k}_{\min} \neq 0$. As it was mentioned above, this case was realized experimentally for the YIG film in Ref.[11], where the critical density of pumped magnons was estimated numerically. Later in Ref.[1] it was considered analytically. In this paper we focus on the second case, where we have just one minimum at $\mathbf{k} = 0$.

The magnon spectrum of the perpendicular magnetized ultra-thin ferromagnetic film can be written as [24]:

$$\omega_k = \left\{ [\omega_H + \omega_{ex}(ak)^2][\omega_H + \omega_{ex}(ak)^2 + \omega_M f(k\tau)] \right\}^{1/2}, \quad (16)$$

where $\gamma = 2\pi \cdot 2.8$ MHz/Oe is the gyromagnetic ratio, $\omega_H = \gamma H_i$, $H_i = H_e - 4\pi M_s + H_{\perp}$ is an effective internal magnetic field, H_{\perp} is the perpendicular anisotropy field, $M_s = 139$ Oe is the saturation magnetization, $\omega_M = 4\pi\gamma M_s = 2\pi \times 4.9$ GHz, $\omega_{ex}a^2 = 2\pi \times 1.09 \times 10^{-2}$ Hz cm² is the exchange constant, $f(k\tau) = 1 - [1 - \exp(-k\tau)]/k\tau$, τ is the thickness of the film, all numerical parameters are given for YIG. Spin waves are assumed to be propagated in the film plane only and there is a uniform magnetization along the depth.

To estimate the critical density of pumped magnons at temperature T , we write the following equation:

$$\frac{N_p}{A_s} = \frac{N(\mu, T)}{A_s} - \frac{N(0, T)}{A_s}, \quad (17)$$

where the sample volume is replaced by the film area A_s . We represent Eq.(17) as

$$\frac{N_p}{A_s} \approx \frac{k_B T}{2\pi} \int_0^{k_T} \left(\frac{1}{\hbar\omega_k - \mu} - \frac{1}{\hbar\omega_k} \right) k dk, \quad (18)$$

This integral at $\mu \rightarrow \hbar\omega_0$ has a logarithmic divergence for an infinitely large film. However, for a finite film we have a magnetostatic mode on the bottom, which can be separated from the spinwave spectrum by a small gap $\Delta\omega$. Thus, we can estimate Eq.(18) as

$$\frac{N_p}{A_s} \approx \frac{k_B T}{4\pi\hbar\omega_{ex}a^2} \ln\left(\frac{\omega_0}{\Delta\omega}\right). \quad (19)$$

Note that this estimate takes into account magnons in the film plane only.

Critical angle

Let us now estimate the critical angle of the magnetic moment deviation from the equilibrium, which is assumed to corresponds to a critical number of excited magnons. This angle is defined by the ratio of perpendicular spin component to its longitudinal component $\tan\theta = S_\perp/S_z$. The perpendicular component is

$$\begin{aligned} S_\perp &= \sqrt{S_x^2 + S_y^2} = \sqrt{\frac{S_+S_- + S_-S_+}{2}} \\ &\approx \sqrt{2Sa^*a} = \sqrt{2SN_p}. \end{aligned} \quad (20)$$

Substituting $S_z \simeq S$, for small angles one obtains

$$\theta \approx \sqrt{\frac{2N_p}{S}} = \sqrt{\frac{2\hbar\gamma}{M_s} \frac{N_p}{A_s\tau}}. \quad (21)$$

For the room temperature $T = 300$ K, film thickness $6 \mu\text{m}$, $\omega_0 = 2\pi \times 2.5$ GHz, $\Delta\omega = 2\pi \times 1$ Hz, we have $\theta_{film} \approx 0.018$. For comparison, an estimate for a bulk sample at the same conditions gives $\theta_{bulk} \approx 0.061$. Taking into account finite thickness of the film, we can expect the experimental value of the angle will be within $\theta_{film} < \theta < \theta_{bulk}$.

Note that in the experiment [25] with thin YIG film, performed by the same method as for nuclear magnon BEC in antiferromagnets [14] and superfluid $^3\text{He-A}$ [9], the authors succeeded to deflect the magnetization on the angle $\theta \approx 0.16$, which is in ten times bigger than a critical angle, calculated in this article. A typical signal for $k = 0$ magnons BEC was observed.

DISCUSSION

In this paper we have focused on critical conditions of BEC in ferro- and antiferromagnets, bulk and thin films. Our recent similar analysis [26] showed a good agreement with experi-

ment for nuclear magnon BEC. Here we have shown that the conditions of BEC formation in one of the most interesting materials, YIG thin film, are fulfilled at small angles, when signals are usually treated as excited spin wave modes. Another very interesting material is hematite, high-temperature antiferromagnet in which the BEC should occur at much lower level of the magnon excitation compared to a ferromagnet.

We did not analyse here a question about stability of magnon Bose-Einstein condensate. The critical density is necessary but not a sufficient condition for uniform magnon BEC. For the BEC stability is important to consider the interaction between magnons [27], and as it was demonstrated in the experiment with superfluid $^3\text{He-A}$ [28], the attractive interaction between magnons destroys condensate even in the case of big magnons density. This problem follows directly from the global minima of Ginzburg-Landau potential. For magnon BEC in ferro- and antiferromagnets this question of condensate stability is still open due to more complicated structure of magnon-magnon interactions.

In conclusion, we emphasize that the Bose-Einstein condensation of quasi-equilibrium magnons is a fundamental law of physics. It cannot be found or simulated using classical Landau-Lifshitz-Gilbert equations. BEC appears due to quantum statistics of quasiparticles in magneto-ordered systems and can exist at room (and even higher) temperatures. One of the most intriguing properties of the BEC is a superfluid spin current, a coherent quantum flow of energy and information. The understanding of magnon BEC in different magnetic materials can be very useful for spin transport and magnonic quantum devices. Interest in the spin currents, the magnetization projection transfer in magnetic materials, is growing every year.

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